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AEROSPACE CORP. EL SEGUNDO CA CHEMISTRY AND PHYSICS LAB F/G 7/4
RATE COEFFICIENTS FOR THE REACTIONS OF BF WITH O AND O₂. (U)

DEC 81 6 C LIGHT, R R HERM, J H MATSUMOTO F04701-81-C-0082

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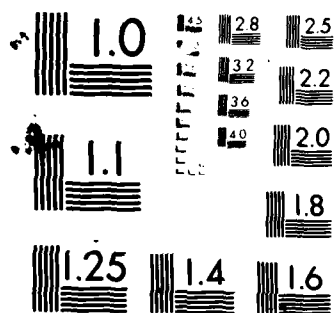
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**Rate Coefficients for the Reactions
of BF with O and O₂**

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Prepared for
SPACE DIVISION
AIR FORCE SYSTEMS COMMAND
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This interim report was submitted by The Aerospace Corporation, El Segundo, CA 90245, under Contract No. F04701-81-C-0082 with the Space Division, Deputy for Technology, P.O. Box 92960, Worldway Postal Center, Los Angeles, CA 90009. It was reviewed and approved for The Aerospace Corporation by S. Feuerstein, Director, Chemistry and Physics Laboratory. Captain David Dise, SD/YLXT, was the project officer for Technology.

This report has been reviewed by the Public Affairs Office (PAS) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER SD-TR-81-101	2. GOVT ACCESSION NO. AD-A109 568	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) RATE COEFFICIENTS FOR THE REACTIONS OF BF WITH O AND O ₂	5. TYPE OF REPORT & PERIOD COVERED Interim	
7. AUTHOR(s) G. C. Light, R. R. Herm, and J. H. Matsumoto	6. PERFORMING ORG. REPORT NUMBER TR-0082(2945-04)-1	
9. PERFORMING ORGANIZATION NAME AND ADDRESS The Aerospace Corporation El Segundo, Calif. 90245	8. CONTRACT OR GRANT NUMBER(s) F04701-81-C-0082	
11. CONTROLLING OFFICE NAME AND ADDRESS Space Division Air Force Systems Command Los Angeles, Calif. 90009	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	12. REPORT DATE 21 December 1981	
	13. NUMBER OF PAGES 15	
	15. SECURITY CLASS. (of this report) Unclassified	
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Boron monofluoride Oxygen molecules Flow tube Reaction rate Gas-phase Room temperature Oxygen atoms		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Bimolecular reaction rate coefficients of $k = 1.4 \pm 0.2 \times 10^{-10}$ and $< 5 \times 10^{-17}$ cm ³ /molecule-sec have been measured at T = 294 K in a flow tube facility for BF + O → BO + F and BF + O ₂ → products, respectively. These results are discussed in terms of the electronic structure of boron monofluoride.		

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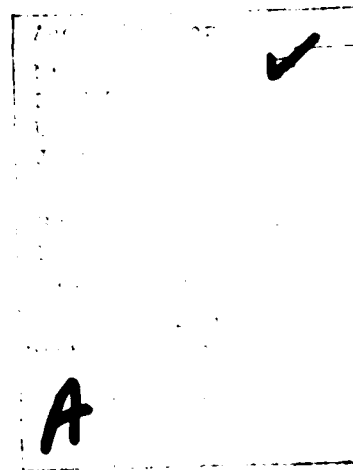
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I. INTRODUCTION

Despite the wide variety of hydride, halide, oxide, oxyhydride, oxyhalide, and other species that can form, there is a scarcity of experimental data available on the rates of reactions of boron compounds. This is especially true^{1,2} for the potentially more interesting compounds wherein the boron atom is formally in a low valence state. The only set of direct kinetic measurements is a recent room-temperature-diffusion flame determination³ of the rates of oxidation of gaseous boron atoms by some inorganic oxides. Other related information is limited to indirect inferences provided by studies of the reaction^{4,5} of oxygen atoms with diborane and the chemiluminescent reactions^{6,7,8,9} of boron atoms. This lack of information is particularly unfortunate because of boron's unique position in the periodic table. The chemistry of metal atoms and their compounds, for example, halides, is often quite different¹⁰ from that of the nonmetals, and boron is the lightest element that divides these two regions of the periodic table. Thus, Parr et al.¹¹ reported

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- ¹S. S. Cherry, L. J. Van Nice, and P. I. Gold, *Pyrodynamics* 6, 275 (1968).
 - ²G. S. Bahn, *Pyrodynamics* 6, 297 (1968).
 - ³U. C. Sridharan, T. G. Diguseppe, D. L. McFadden, and P. Davidovits, *J. Chem. Phys.* 70, 5422 (1979).
 - ⁴C. W. Hand and L. K. Derr, *Inorg. Chem.* 13, 339 (1974).
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 - ⁶S. P. Tang, N. G. Utterback, and J. F. Friichtenicht, *J. Chem. Phys.* 64, 3833 (1976).
 - ⁷U. C. Shridharan, D. L. McFadden, and P. Davidovits, *J. Chem. Phys.* 65, 5373 (1976).
 - ⁸A. Brzychcy, J. Dehaven, A. T. Prengel, and P. Davidovits, *Chem. Phys. Lett.* 60, 102 (1978).
 - ⁹G. J. Green and J. L. Gole, "Contrasting Chemiluminescent Metal Oxide Formation in the Single Collision Reactions of Boron with O₂, NO₂, N₂O, O₃, and ClO₂," *Chem. Phys. Lett.* 69 (1), 45 (1980).
 - ¹⁰R. R. Herm, "Reactive Scattering of Alkali Atoms and Alkali Halides at Thermal Energies," in *Alkali Halide Vapors. Structure, Spectra, and Reaction Dynamics*, eds., P. Davidovits and D. L. McFadden, Academic Press, New York (1979).
 - ¹¹T. P. Parr, R. Behrens Jr., A. Freedman, and R. R. Herm, *Chem. Phys. Lett.* 56, 71 (1978).

between metallic and nonmetallic reaction dynamics occurs somewhere between tin and mercury; the ionization potentials of tin, boron, and mercury are 7.3, 8.3, and 10.4 eV, respectively. Indeed, Green and Gole⁹ found that the language of the familiar electron transfer model is useful in discussing their observations on boron chemiluminescent reactions.

This is a report of the first results from our laboratory of a flow-tube study of some reaction rates of boron monofluoride, BF. This molecule is capable of very exothermic reactions with a number of reagents; exoergicities from the JANAF Tables for the



and



reactions, where X denotes an inert third body, are provided in Table 1. However, very little is known about the chemistry of BF. Moeller and Silvers¹² produced BF by a microwave discharge in BF₃ and reported that it was surprisingly long-lived in their flow apparatus when mixed with helium. Thermodynamic calculations indicate that BF₃ gas should be quantitatively converted to BF gas upon passage over hot boron powder. Indeed, the heat of formation and entropy of gaseous BF were first measured by this transpiration technique,¹³ and it was subsequently employed by Timms^{14,15} in one of the pioneering studies of the synthesis of new compounds by co-condensation of a high-temperature

¹²M. B. Moeller and S. J. Silvers, Chem. Phys. Lett. 19, 78 (1973).

¹³J. Blauer, M. A. Greenbaum, and M. Farber, J. Phys. Chem. 68, 2332 (1964).

¹⁴P. L. Timms, J. Am. Chem. Soc. 89, 1629 (1967).

¹⁵P. L. Timms, Adv. Inorg. Chem. Radiochem. 14, 121 (1972).

Table 1. Reaction Energetics and Rates^a

Reaction	Exoergicity, ΔH_0°	Rate Coefficient ^b	
		$\log_{10} A$	E^*
$\text{BF} + \text{O} \rightarrow \text{BO} + \text{F}$	-54	-9.9	~0
$\text{CO} + \text{O} \rightarrow \text{CO} + \text{O}$	0	-10.0	29
$\text{N}_2 + \text{O} \rightarrow \text{NO} + \text{N}$	+314	-9.9	320
$\text{BF} + \text{O} + \text{X} \rightarrow \text{OBF} + \text{X}$	-730	-	-
$\text{CO} + \text{O} + \text{X} \rightarrow \text{CO}_2 + \text{X}$	-526	-	-
$\text{BF} + \text{O}_2 \rightarrow \text{OBF} + \text{O}$	-237	-	-
$\quad \quad \quad \rightarrow \text{BO}_2 + \text{F}$	-89	-	-
$\text{CO} + \text{O}_2 \rightarrow \text{CO}_2 + \text{O}$	-33	-11.4	200
$\text{N}_2 + \text{O}_2 \rightarrow \text{N}_2\text{O} + \text{O}$	+332	-10.0	460
^a Energies in kJ/mole. ^b Parameters of the bimolecular rate coefficient: $k(\text{cm}^3/\text{molecule-sec}) = A \exp[-E^*/RT]$			

vapor with another reagent onto a cryogenic surface. Timms^{14,15} found that the useful synthesis reactions of BF were limited to insertion into other B-F bonds and addition across double and triple bonds in alkenes and alkynes.

II. EXPERIMENTAL PROCEDURE AND RESULTS

Full details of the apparatus and experimental procedure will be published later. Briefly, rate coefficients for Reactions (1) and (3) have been measured at 294 K in the 2.5-cm diameter flow tube shown in Fig. 1. The reaction zone, defined at the upstream and downstream ends by the regions where reactants were mixed and BF was detected, respectively, was of fixed length, $l = 87.5$ cm.

The BF was detected by resonance fluorescence at 90 degrees to the BF resonance excitation light, which was provided by the output of a microwave discharge in BF_3 similar to that described in Ref. 12. The $\Delta v = 0$ $A^1\pi \rightarrow X^1\Sigma^+$ BF emission band at 195.6 nm was isolated by a filter that rejected other significant lamp outputs at 191, 201, and 207 nm, from $\Delta v = +1, -1$, and -2 transitions of this same BF system, and at 208.9 and 249.7 nm from boron atomic lines.

The BF was generated in a manner similar to that described in Refs. 13 and 14 by flowing BF_3 vapor in an argon carrier through a vacuum enclosed resistively heated graphite tube at >1800 K; the tube was packed with 99.8% purity crystalline boron granules. A General Electric inert-gas purifier reduced the oxygen and water impurities to less than 0.1 ppm in the Matheson UHP argon. The effluent from the furnace flowed through a quartz tube (diameter, 1.0 cm; length, 51 cm) into the main flow tube where it had cooled to room temperature and was mixed with more carrier gas. If thermodynamic equilibrium were achieved within the furnace, BF would represent $>99.9\%$ of the boron-containing effluent because of the



reaction. The actual BF concentration entering the flow tube was less than this value, which is denoted $[\text{BF}]_{\text{max}}$ because of incomplete equilibrium within the furnace or reactions during the transit from the furnace to the flow tube. This was inconsequential, however, because relative, rather than absolute, BF

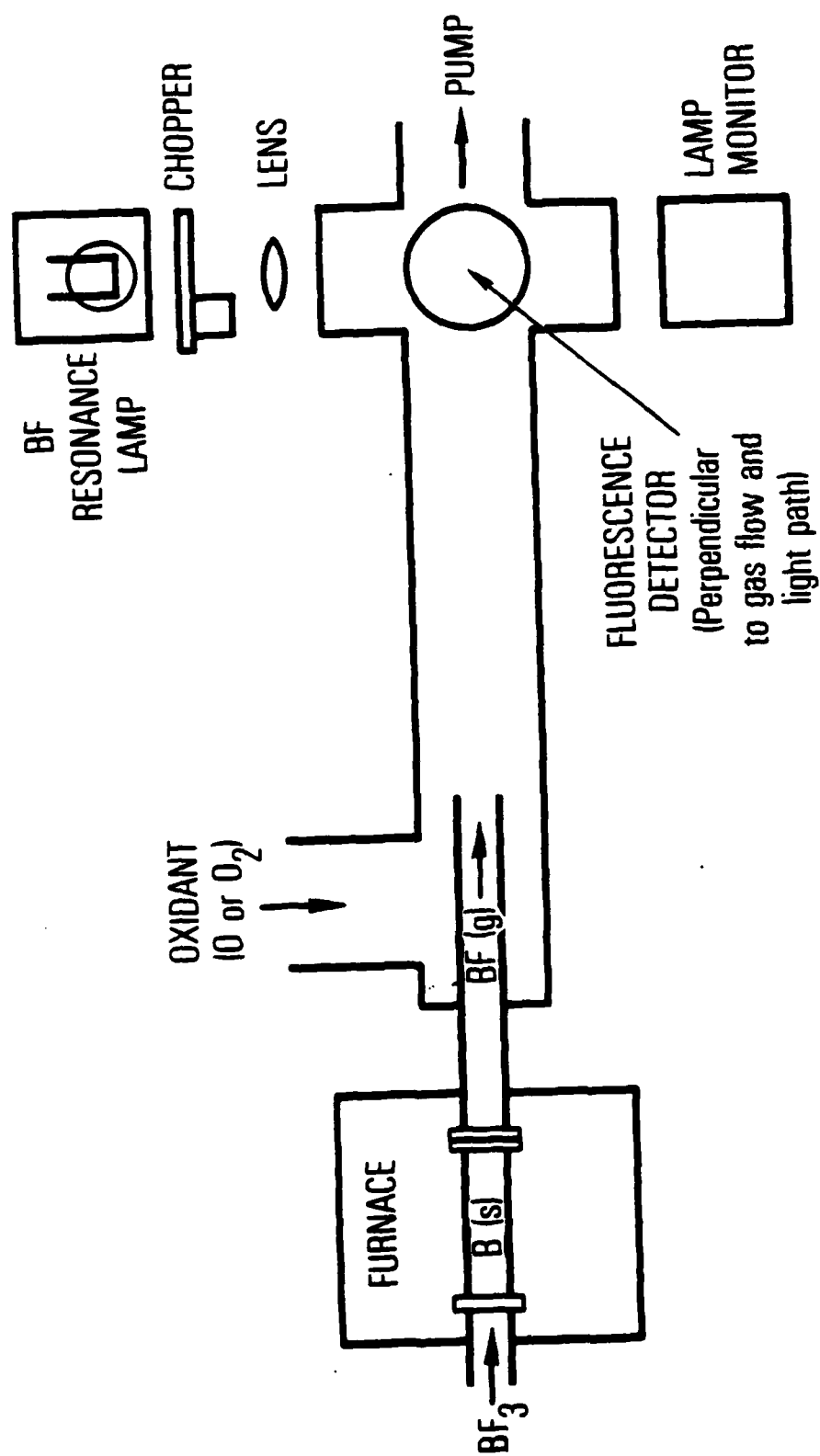


Figure 1. Flow Tube Facility

density measurements were required for the kinetics study. The total density of boron species within the flow tube, controlled by measuring and limiting the BF_3 flow rate into the furnace, was always sufficiently low to ensure negligible probability of reaction between two boron species within the reaction zone. Since other boron species could not react with any other reagent to generate BF, this ensured the total insensitivity of the BF density to the possible presence of any other boron species. This limit on the maximum BF flow tube density also ensured optically thin conditions for the BF resonance fluorescence detection. Since collisional quenching of the $A^1\pi$ state of BF was negligible because of its short radiative lifetime, 2.8 nsec,¹⁶ proportionality of the BF density within the flow tube to the detected resonance fluorescence was ensured.

This also ensured that the BF density was always much less than that of the O or O_2 co-reactant so that pseudo-first order decay of BF was achieved; that is, the BF density at the detector depended upon O or O_2 density as

$$\ln \frac{[\text{BF}]}{[\text{BF}]_0} = - \frac{k l}{\langle v \rangle} \quad (5)$$

where $k = k_1[\text{O}]$ or $k_3[\text{O}_2]$ in the study of Reactions (1) and (3), respectively, and $\langle v \rangle$ is the average flow velocity, calculated from the mass flow rate of the overwhelmingly abundant argon carrier gas and the measured total flow tube pressure. Typically, the total pressure was 2 to 9 Torr with $\langle v \rangle \sim 1400$ cm/sec; for these conditions, the fractional pressure drop over the reaction zone was less than 5%. The assumption of plug flow implied in Eq. (5) is a good approximation for the data reported herein; small corrections for deviation from plug flow will be included in a more comprehensive report.

Admission of O_2 to a flowing BF-Ar mixture had no effect on the BF density detected even at the highest O_2 density employed, $7 \times 10^{15} \text{ cm}^{-3}$. Thus, the room-temperature rate of Reaction (3) was too slow to measure, and an upper limit for the rate coefficient was established:

¹⁶ T. E. Esser, J. Chem. Phys. 48, 2518 (1968).

$$k_3 = k_{3a} + k_{3b} < 5 \times 10^{-17} \text{ cm}^3/\text{molecule-sec} \quad (6)$$

Addition of ground state ^3P O-atoms, however, rapidly attenuated the BF signal due to Reaction (1). The $\text{O}(^3\text{P})$ density was quantitatively varied by the well-known



titration technique after ascertaining that addition of active nitrogen itself had no influence on the BF density for the experimental conditions employed. Details of this technique are provided in Ref. 17. Figure 2 is representative of the data obtained for Reaction (1). Note that this set of data demonstrates that the rate is first order in BF and O densities over factors of ~ 90 and ~ 5 , respectively. Similar data were taken for a variety of flow tube pressures and BF_3 flow rates; the results are provided in Table 2. The final rate coefficient obtained for Reaction (1) at 294 K by averaging the results of all of these experiments is

$$k_1 = 1.4 \pm 0.2 \times 10^{-10} \text{ cm}^3/\text{molecule-sec} \quad (8)$$

The very fast rate of Reaction (1) precluded the collection of any data on the termolecular Reaction (2).

¹⁷G. C. Light, J. Chem. Phys. 68, 2831 (1978).

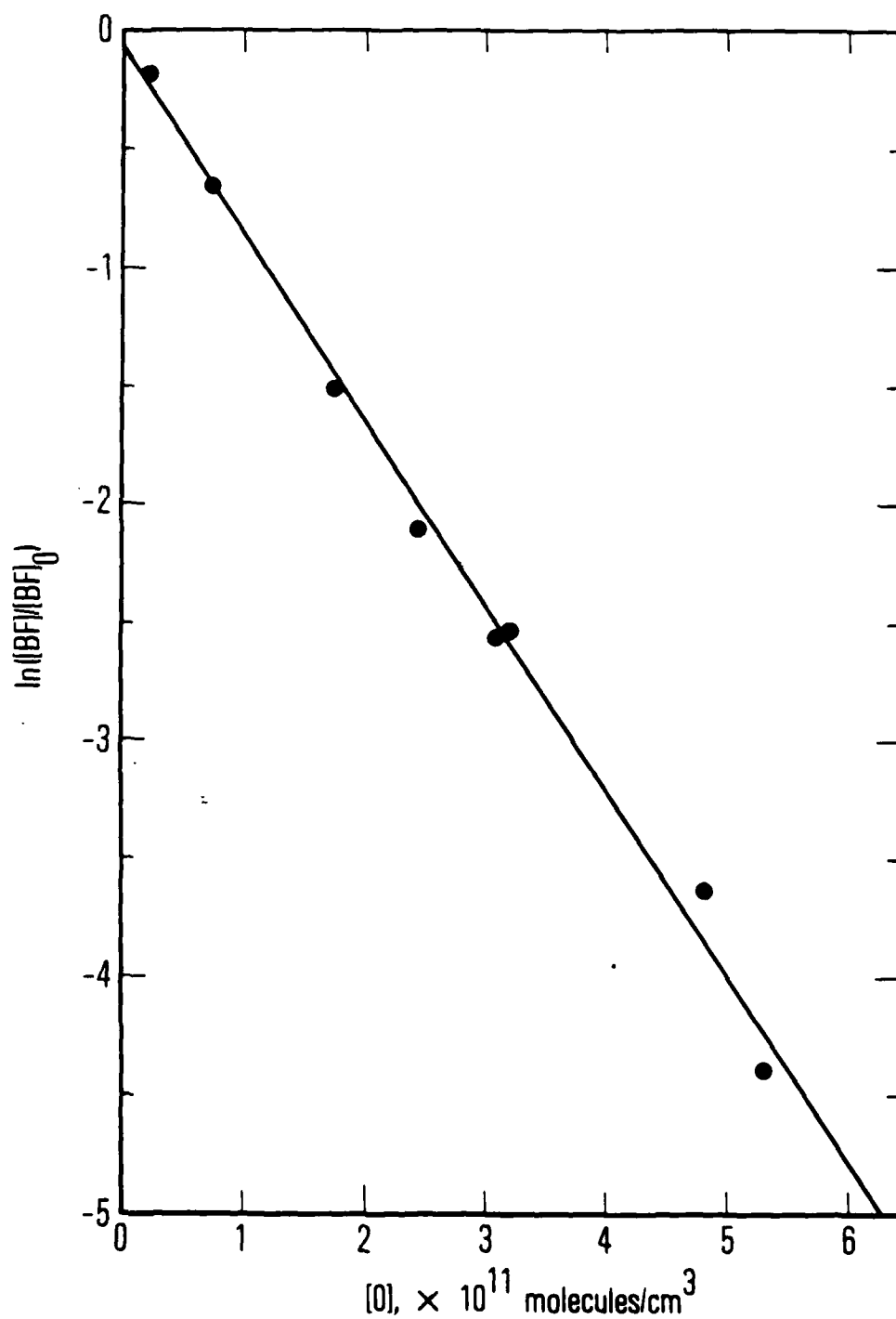


Figure 2. Natural Logarithm of the Measured Ratio of BF Densities With and Without O-Atoms Added Versus O-Atom Density for Typical $\text{BF} + \text{O} \rightarrow \text{BO} + \text{F}$ Experiment. Solid line demonstrates the fit of Eq. (5) to the data.

Table 2. Summary^a of Separate Studies of Reaction (1)

Total Pressure, Torr	[BF] _{max} , 10 ¹¹ cm ⁻³	k ₁ , 10 ⁻¹⁰ cm ³ /molecule-sec
2.9	1.0	1.20
2.9	4.0	1.27
2.8	6.2	1.49
8.7	4.9	1.26
2.3	b	1.58
2.8	3.4	1.59
2.8	3.3	1.66
^a [BF] _{max} is the concentration of BF at the upstream end of the reaction zone calculated from the measured BF ₃ flow rate, assuming that Reaction (4) went to completion. k ₁ is calculated by fitting Eq. (5) to the data. ^b Not measured but comparable to the other entries.		

III. DISCUSSION

A comparison of our measured rates of Reaction (1) and the rates of the reactions of isoelectronic N_2 and CO is provided in Table 1. These rates for N_2 and CO are taken from the recent review of Ref. 18 except for the rate of the $O + CO$ isotopic exchange reported in Ref. 19. Some properties, such as the unusually large bond dissociation energies, support a simple isoelectronic projection of comparable behaviors for these three molecules. This is further supported by the small polarity of the BF bond, a dipole moment²⁰ of only $0.5 \pm 0.2D$. However, other considerations suggest that the chemistry of BF might be quite different from that of CO and N_2 . Quite different exoergicities for formally analogous reactions of these species are indicated in Table 1. Moreover, electronic structure calculations^{21,22} indicate that individual molecular orbitals of BF are quite asymmetric so that the small overall dipole moment could be misleading. Nevertheless, it is appropriate in discussing our results on the previously unexplored chemical behavior of BF to employ the simplest consistent model, and the analogy to the isoelectronic N_2 and CO is useful in interpreting the present observations. It will be interesting to determine whether these isoelectronic analogies continue to be useful as more detailed data on BF reactions are obtained.

The $BF + O$ reaction rate coefficient is quite large, indicating a zero or very small activation energy and a large average total reaction cross section, $\langle \sigma \rangle = k/\langle g \rangle = 19 \text{ \AA}^2$. This is similar to the behavior of the N_2 and $CO + O$ rates in Table 1, namely, preexponential factors quite close to that found for BF, and activation energies only slightly larger than the endoergicity.

¹⁸R. F. Hampson Jr. and D. Garvin, Reaction Rate and Photochemical Data for Atmospheric Chemistry 1977, NBS Special Publication 513, U.S. Department of Commerce, Washington, D.C. (May 1978).

¹⁹S. Jaffe and F. S. Klein, *Trans. Faraday Soc.* **62**, 3135 (1966).

²⁰F. J. Lovas and D. R. Johnson, *J. Chem. Phys.* **55**, 41 (1971).

²¹P. Sutton, P. Bertoncini, G. Das, T. L. Gilbert, A. C. Wahl, and O. Sinanoglu, *Int. J. Quantum Chem.* **3**, 479 (1970).

²²H. B. Jansen and P. Ros, *Theor. Chim. Acta (Berlin)* **21**, 199 (1971).

Moreover, a survey of the rate coefficients for a variety of O-atom reactions given in Ref. 18 indicates that these three quite similar preexponential factors are unusually large. The nature of the relevant potential energy surfaces for $O + CO$ is discussed in Refs. 23 and 24. The small 29-kJ/mole activation energy apparently arises because the initial $O + CO$ triplet potential surfaces are repulsive since they correlate to higher energy states of the CO_2 intermediate. However, they are crossed by triplet surfaces correlating to CO_2 states that are bound or only slightly repulsive with respect to separated reactants so that only a small barrier results. The absence of any barrier for the analogous BF reaction found in this study is consistent with the larger exoergicity of Reaction (2) for $BF + O$ (Table 1). This should shift the surface crossing to larger reactant separation and also reduce or eliminate the repulsion of the initial $O + BF$ potential surfaces by lowering the energy of the OBF states to which they correlate.

The limited information obtained here on the $BF + O_2$ reaction is also consistent with the behavior of the CO and $N_2 + O_2$ reactions. Temperature dependent measurements of the rate of Reaction (3), which are now being made, will permit a more quantitative comparison of these three reactions.

²³J. D. Kelley and R. L. Thommarson, J. Chem. Phys. 66, 1953 (1977).

²⁴J. E. Hardy, W. C. Gardiner Jr., and A. Burcat, Int. J. Chem. Kinet. 10, 503 (1978).

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